Presentation to Brookhaven Workshop 9th and 10th May 2007

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The Development of Assay Procedures for WAGR Decommissioning Waste

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WAGR Waste Assay

- The early calculations used to asses the dose rates for decommissioning highlighted the sensitivity of a calculated inventory to the neutron flux in a particular region, the elemental levels of the precursors, and the neutron capture cross sections of these precursors.

- It was clear that a more detailed radio-isotopic inventory would be required for transport and disposal purposes covering the difficult to measure low energy gamma and pure beta emitters and very long half life isotopes which are important for long term storage.

- Programmes of work both nationally and internationally to establish the best available cross sectional data accelerated around this time.

- Searches for archive material were put in hand and where possible limited programmes of sampling and analysis were carried out to fill in missing material compositional data.

WAGR Assay – Real time material sampling and analysis?

- At this time it was decided that information on several important trace elements was either inadequate or not available and that at the time of decommissioning sampling and analysis of material from each box load of waste would be needed to meet foreseen regulatory requirements.
- Each sample taken would be split into three. Two would go for analysis and the third would be archived.
- Because of the need to turn the samples sent for analysis round rapidly so as not to disrupt the dismantling process until the samples were returned and the box contents shown to satisfy regulatory requirements, the decision was taken to set up a local analytical laboratory purely to analyse WAGR material specimens.
- Work was put in hand to establish suitable analytical processes, acquire the appropriate analytical equipment and find suitable local premises.

Continued -

WAGR Assay – Real time material sampling and analysis?

- Premises were found adjacent to the reactor building, and much of the required analytical equipment, fume hoods and shielded handling facilities purchased and installed.

- In parallel with this work options for designs and procedures for a radioactive material archive store were studied, and suitable shielded facilities investigated.

- In reality none of the analytical equipment was ever used – the lab never went active and the concept of using an archive store was abandoned – it was all too expensive and the level of inventory accuracy needed did not require it. The regulators had never really been asked before about the level of accuracy they required.

Further Inventory Modelling

- Detailed modelling work on the core using the ANISN neutron transport code created a radioactive inventory for all major items in the core for a list of isotopes deemed to be greater than that required by regulators.
- Further material sampling campaigns had improved the available compositional data which were used for this modelling.
- The code used 22 energy group cross sectional data set to calculate axial and radial flux distributions.
- The flux distributions created were then used in neutron activation codes to generate the inventories.

Nuclear reactions considered for modelling purposes

Parent	Reaction	Daughter	Half life (y)	Abundance
Li-6	n,alpha	H-3	12.3	7.5
C-13	n,gamma	C-14	5.73×10^3	1.1
N-14	n,p	C-14	5.73×10^3	99.6
Cl-35	n,gamma	Cl-36	3.01×10^5	75.77
Ca-40	n,gamma	Ca-41	1.0×10^5	96.94
Fe-54	n,p	Mn-54	0.855	5.8
Mn-55	n,2n	Mn-54	0.855	100
Fe-54	n,gamma	Fe-55	2.7	5.8
Ni-58	n,gamma	Ni-59	7.5×10^4	68.27
Ni-62	n,gamma	Ni-63	96	3.59
Co-59	n,gamma	Co-60	5.272	100
Nb-93	n,gamma	Nb-94	2.0×10^4	100
Mo-94	n,p	Nb-94	2.0×10^4	9.25
Nb-93	n,n'gamma	Nb-93m	13.6	100
Eu-151	n,gamma	Eu-152	13.4	47.8
Eu-152	n,gamma	Eu-153	Stable	0
Eu-153	n,gamma	Eu-154	8.5	52.2
Eu-154	n,gamma	Eu-155	4.73	0

WAGR Waste Assay

- The later sampling and analysis campaigns had included isotopic measurements on activated material and this information was available for comparison with calculated values.

- The table on the next few slides provides some comparisons of the calculated activities with the measured values.

Comparison of calculated and measured values (C/M)

Graphite core					
Position	Co-60	H-3	C-14	Eu-154	Eu-152
Upper core	0.72			33.7	
Upper core	0.9			6.1	
Upper core	0.056	2.97	1.29		
Upper core	0.24	0.88	2.59		

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Comparison of calculated / measured values

Loop tube stainless steel					
Position	Co-60	H-3	C-14	Eu-154	Eu-152
Hot box region	0.21				
Neutron shield	1.64				
Upper core	1.56				
Mid core	1.64				

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Comparison of calculated / measured values

Pressure vessel					
Position	Co-60	H-3	C-14	Eu-154	Eu-152
Mid core	2.04				

Pressure vessel outer insulation wire					
Position	Co-60	H-3	C-14	Eu-154	Eu-152
Mid core	1.47				

Pressure vessel insulation					
Position	Co-60	H-3	C-14	Eu-154	Eu-152
Mid core		6.5			
Mid core		6			
Mid core		1.1			

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Comparison of calculated / measured values

Ventilation membrane					
Position	Co-60	H-3	C-14	Eu-154	Eu-152
Mid core	1.36				

Bioshield re-bar					
Position	Co-60	H-3	C-14	Eu-154	Eu-152
Mid core	2.67				
Mid core	2.03				

Bioshield concrete inner face					
Position	Co-60	H-3	C-14	Eu-154	Eu-152
Mid core	1.02	1.61		2.22	1.59

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Calculated average core fluxes

Neutron energy band	Thermal 0.0001eV to 0.625eV	Resonance 0.625eV to 9.1keV	Fast 9.1keV to 10MeV
Averaged flux ($n/cm^2/s$)	0.79×10^{13}	1.34×10^{13}	0.79×10^{13}

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WAGR Waste Assay Procedures

- **The modelling and sampling and analysis work has shown:**
  - There is a wide spread in the analytical results particularly for the trace elements at the low ppm and ppb levels.
  - The quality of currently available neutron cross-sectional data allows modelling using neutron transport theory to compute flux spectra which can be used in activation codes to generate data in which the main source of error comes from the the parental level uncertainties.

WAGR Waste Assay Procedures

- This leads to the possibility of using modelling to provide the relative levels of the isotopes which need recording in the inventory.

- The absolute levels however do need tying down.

WAGR Waste Assay Procedures

- WAGR has essentially only two types of waste material – steel and graphite.
- The bulk of the steel is mild steel, coming from the thermal shield and the pressure vessel.
- There are also reasonable quantities of stainless steel (type 321 and 347) and boronated steels.
- The predominant gamma ray emitter and hence dose rate generator is Co-60.
- There is some fission product contamination, the majority of which ended up in the hot box and heat exchangers which both acted as a very good filters.
- Cs137 is the main detectable fission product at this time.

WAGR Waste Assay Procedures

- It was decided that Co-60 should be used as the scaling isotope.
- Because it was the prime gamma emitter, dose rate alone could be used to establish Container Co-60 inventory.
- Software was developed and tested which allowed multiple dose point dose rates from around a waste container to be used in an inverse shielding computation to establish the Co-60 inventory of the container.
- The software used the waste mass in the container to allow for self shielding effects.

WAGR Waste Assay Procedures

- To cater for taking dose rate measurements around a waste container a set of four ion chambers on a 1m meter square array were provided in the lower loading cell.
- The ion chambers were mounted on plungers which allow dose point positions to be adjusted to suit the waste form.
- The layout is shown in the next slide

The Upper Loading Cell Assay Station

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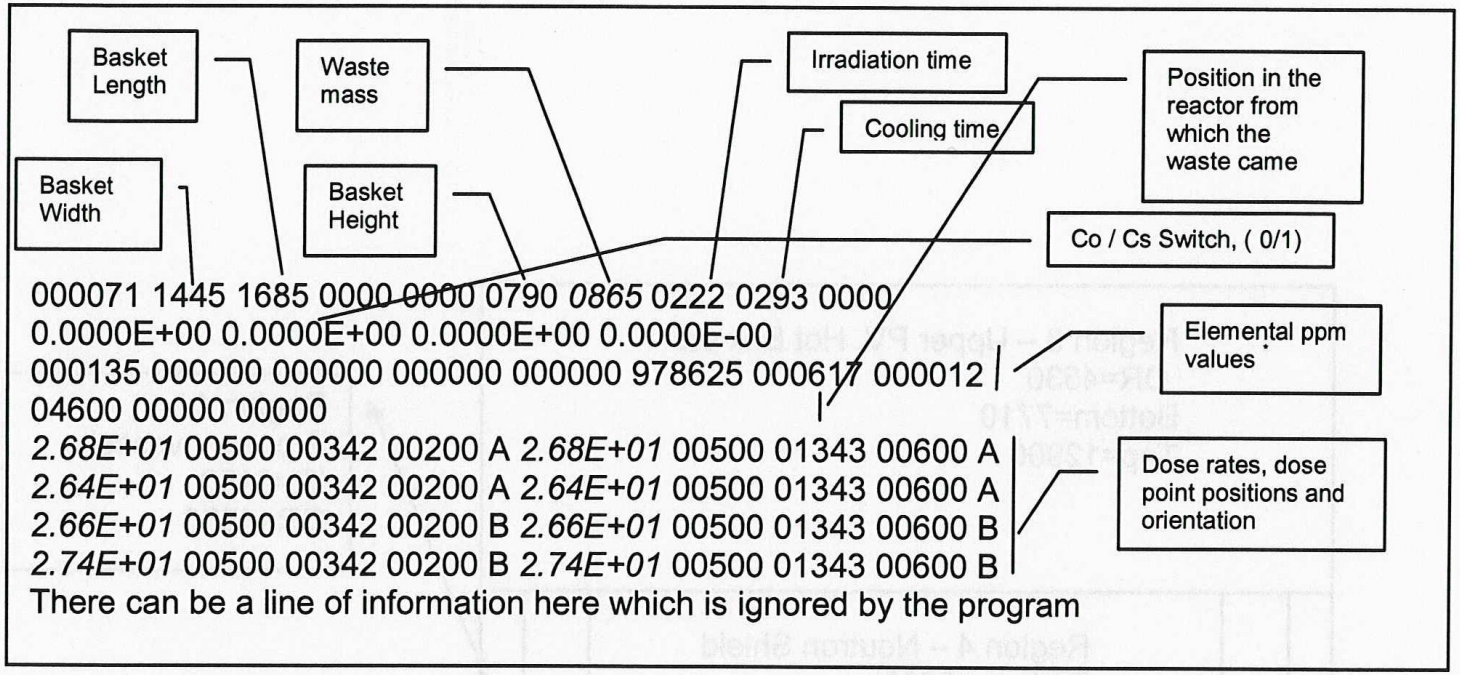
WAGR Waste Assay Procedures

- The software was subsequently extended to use parental level of Co-59 and its capture cross section to calculate the integrated neutron flux required to convert the Co-59 to the observed quantity of Co-60.

- Having obtained the neutron flux, the levels of the other isotopes required for the inventory could be calculated using their parental levels and capture cross sections.

- The next slide shows the input data required by the code.

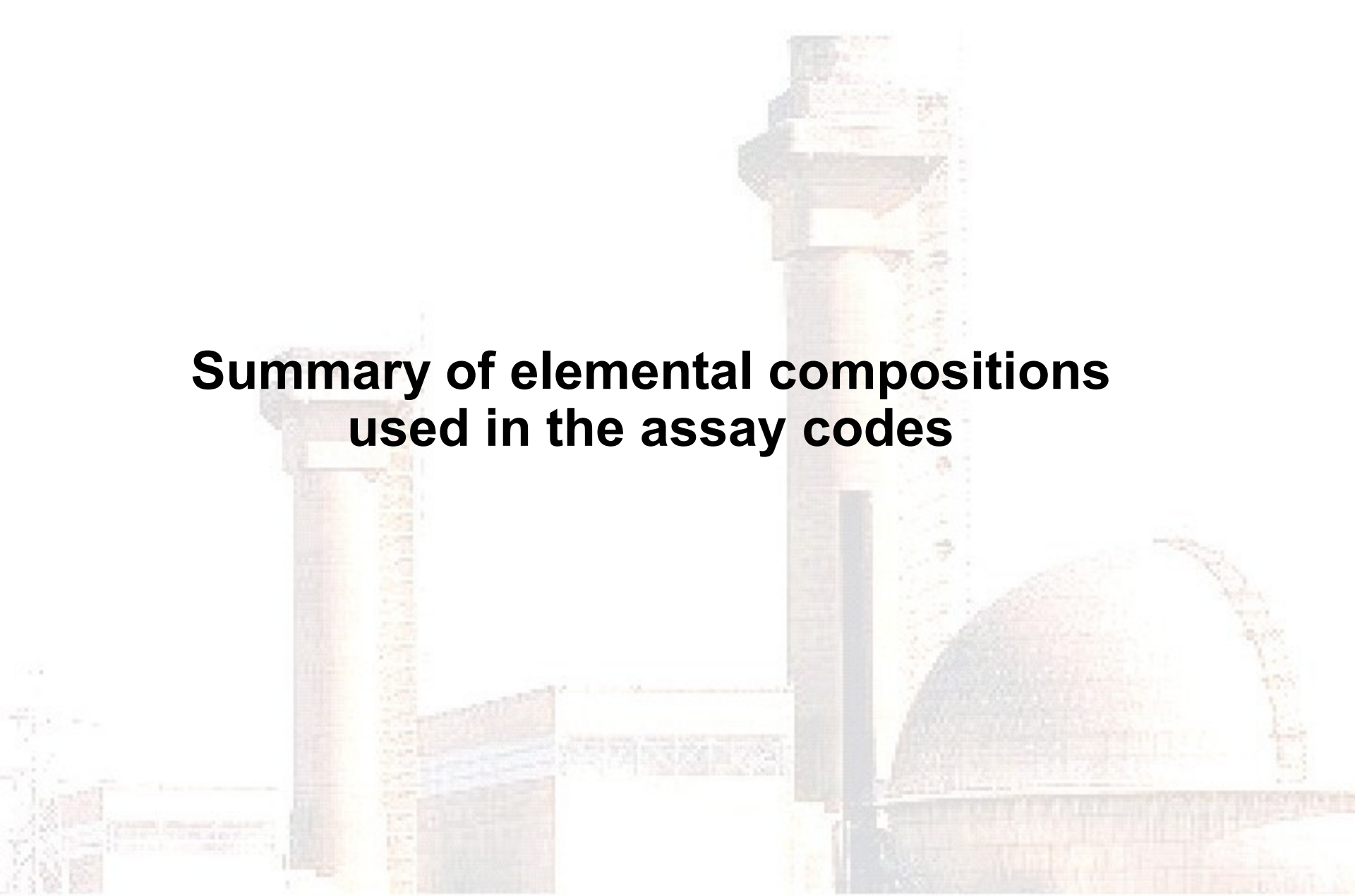


* In the order C0, Li, N, Cl, a spare, Fe, Ni, Nb

The formats in the file must be adhered to rigidly or the case will not run correctly.

The A or B character at the end of each set of four values representing dose rate and (x,y,z) dose point position realative to the bottom left hand corner of the waste are to assist with the input of data. When the B is present the code assumes that the waste has been rotated through 180° allowing the user to give (x,y,z) values relative to the new bottom left hand corner rather than having to transform the co-ordinates by rotation and transalation to the old pre-rotation corner.

Summary of elemental compositions used in the assay codes



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Type 321 Stainless Steel

Parental elemental concentrations			
Co	517	ppm	+/-274
Li	0	ppm	
Eu	0	ppm	
Cl	0	ppm	
Fe	610350	ppm	+/-56616
Ni	107385	ppm	+/-10048
Nb	166	ppm	+/-84

Type 347 Stainless Steel

Parental elemental concentrations			
Co	1467	ppm	+/-864
Li	0	ppm	
Eu	0	ppm	
Cl	0	ppm	
Fe	676333	ppm	+/-3512
Ni	98500	ppm	+/-8698
Nb	5740	ppm	+/-4483

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Mild Steel

Parental elemental concentrations			
Co	135	ppm	+/-55
Li	0	ppm	
Eu	0	ppm	
Cl	0	ppm	
Fe	978625	ppm	+/-7891
Ni	617	ppm	+/-280
Nb	12	ppm	1 value

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Graphite

Parental elemental concentrations			
Co	11	ppb	+/-3
Li	343	ppb	+447/-343
Eu	6	ppb	+9/-6
Cl	3	ppm	+/-3
Fe	3	ppm	+/-1
Ni	1	ppm	+/-0.4
Nb	0	ppm	?

The Assay codes

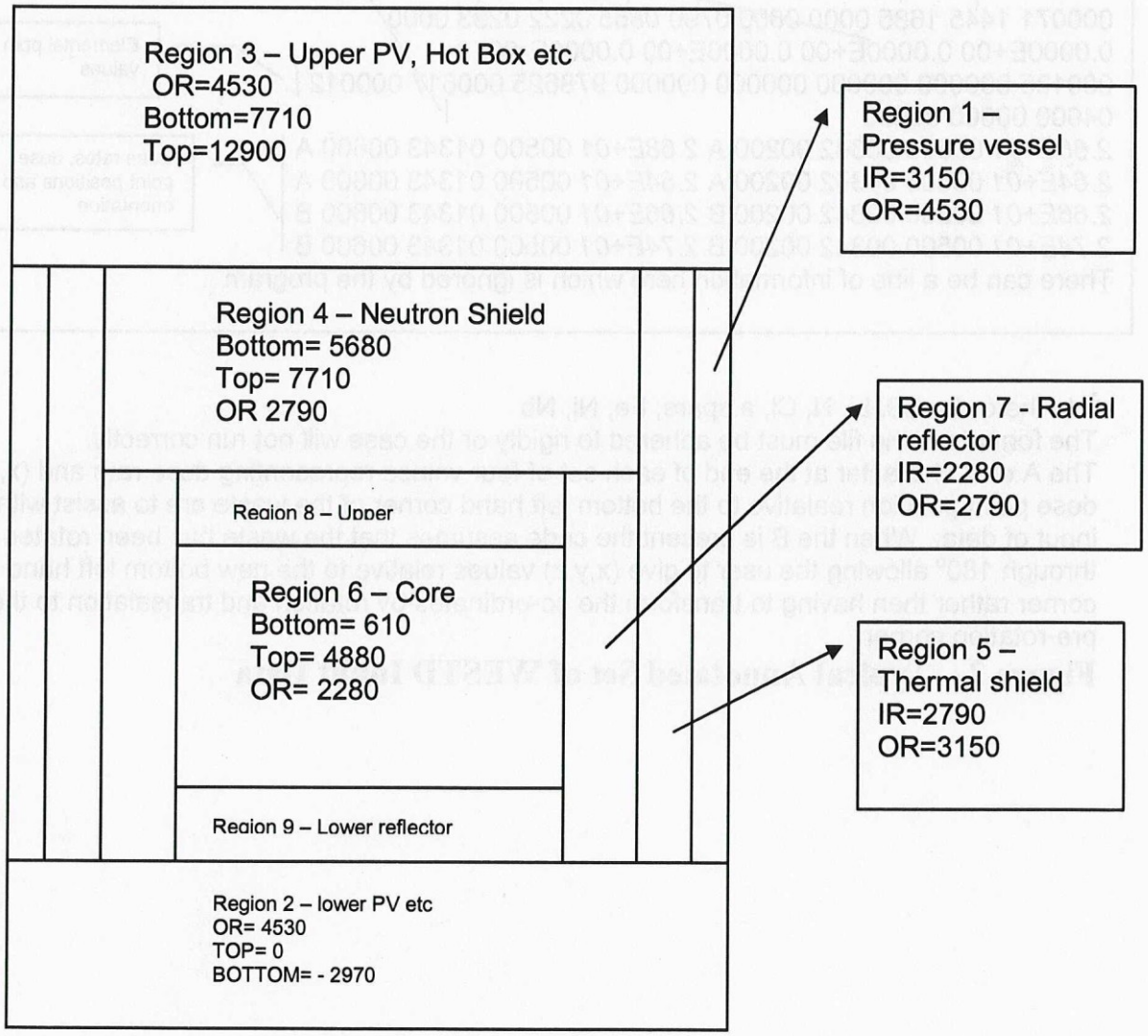
- There are two potentially complicating factors which arise when assessing the inventory by these means.
- Firstly the variation of neutron capture cross section varies with neutron energy, and the neutron energy spectrum varies from point to point in the reactor.
- Secondly the generation of Co-60 from Co-59 is a function of three competing processes:
  - An increase as the parental Co-59 absorbs neutrons to generate Co-60
  - A decrease as the Co-60 decays
  - A decrease in the generation rate from Co-59 as it gets depleted

  The Co-60 level hence rises until the decay matches the falling rate of production and then starts to fall. Hence a given Co=60 inventory can be generated by two levels of irradiation.

The Assay codes

- To overcome the first problem the reactor was divided into regions and sets of condensed cross sections and neutron flux spectra generated for each region.

- The regions used are shown in the next slide.

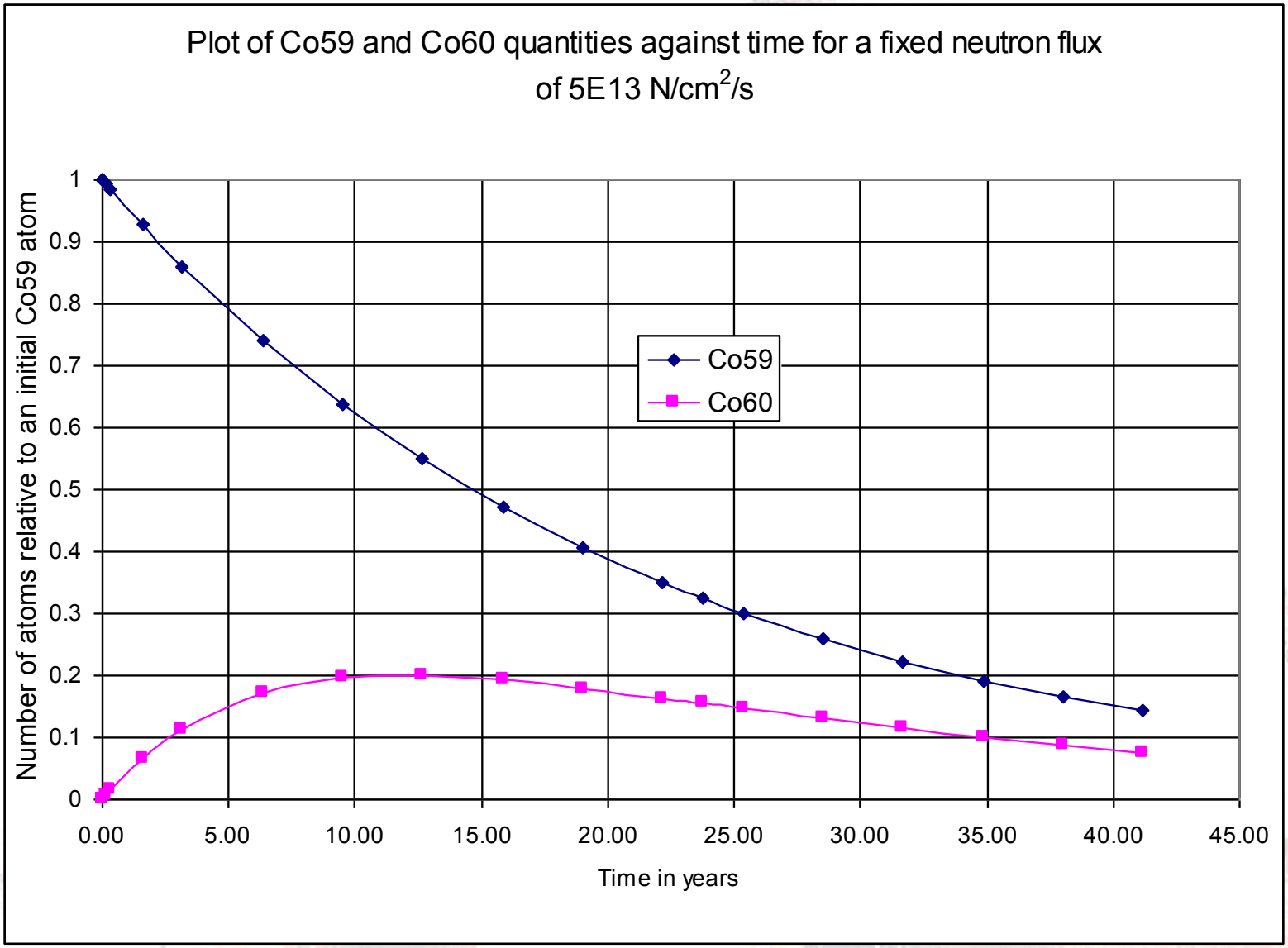


Region 10 covers all space external to the above

The Assay codes

- As regards the second point the figure on the next slide shows the behaviour of Co-59 and Co-60 for a 1 group neutron flux of 5×10^{13} n/cm^2/s. The Co-60 level peaks at around 12 years.

- Fortunately the peak neutron flux in the WAGR was only 1.5×10^{13} n/cm^2/s. The peak occurs at ~19 years irradiation and the problem does not arise (see the subsequent slide).


Plot of Co59 and Co60 quantities against time for a fixed neutron flux of 5E13 N/cm2/s
Number of atoms relative to an initial Co59 atom
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
0.00
5.00
10.00
15.00
20.00
25.00
30.00
35.00
40.00
45.00
Time in years
Co59
Co60

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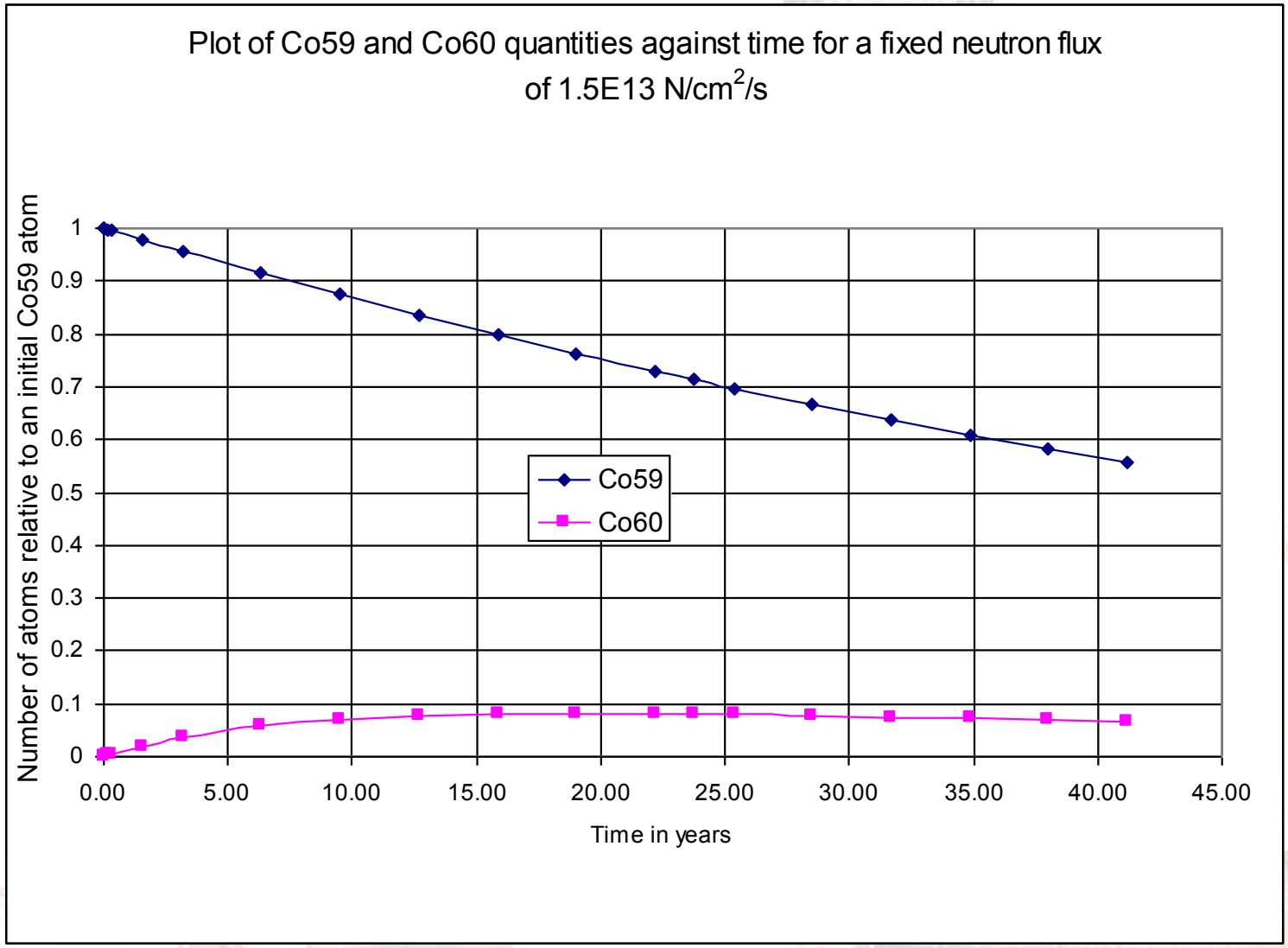
Plot of Co59 and Co60 quantities against time for a fixed neutron flux of 1.5E13 N/cm2/s
Number of atoms relative to an initial Co59 atom
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
0.00
5.00
10.00
15.00
20.00
25.00
30.00
35.00
40.00
45.00
Time in years
Co59
Co60

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The Assay codes

- Four variants of assay code were developed which are generally referred to as the WEST codes.
- WESTD For rectangular baskets of miscellaneous waste.
- WESTE For rectangular plate sections such as slabs of thermal shield and the pressure vessel.
- WESTF For tube sections – used for operational waste such as control rods, neutron shield plugs, arrestor mechanisms and specimen containers.
- WESTG Specially made to handle the grouted, coaxial loop tube sections.

The Assay codes

- Certain reactor components were exposed to the coolant stream during reactor operations and could be surface contaminated with Cs-137.
- The Cs-137 would contribute to the external dose rate.
- A High Resolution Gamma Spectrometer (HGRS) viewing the waste through a collimating hole in the Upper Loading Cell wall had been installed to allow the contribution to dose rate from Cs-137 seen by the ion chambers to be measured and subtracted to obtain the Co-60 contribution. The HGRS analytical software reported the maximum level of Cs-137 which could be present but would not be detected.
- The WEST codes were also capable of using the dose rate from Cs-137 alone to provide the Cs-137 inventory in the waste package.

Typical Output from the WESTD code used for miscellaneous steel waste in a rectangular basket

```
 Estimate that PHI = 1.4483E+11
**************************************
CO60   Activity is 4.0833E+02 GBq.
CS137  Activity is 0.0000E+00 GBq.
H3     Activity is 0.0000E+00 GBq.
C14    Activity is 0.0000E+00 GBq.
CL36   Activity is 0.0000E+00 GBq.
FE55   Activity is 2.0570E+02 GBq.
NI63   Activity is 2.1305E+03 GBq.
NB94   Activity is 1.3154E+00 GBq.
Total  Activity is 2.7459E+03 GBq.
**************************************
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Assay of WAGR Graphite

- At the time that the graphite components were boxed and grouted no confirmatory assay checks were performed.

- The inventories used were based on the isotopic levels generated by the later modelling work.

Assay of WAGR Graphite

- The next two slides give the elemental composition of the graphite used in the modelling work and the actual Inventory data used for the packaged graphite obtained by multiplying the mass of the graphite by the Bq/g for the originating location.

Elemental make up of graphite used for modelling purposes

Element	ppm	Element	ppm
Boron	0.096	Molybdenum	0.062
Cadmium	0.04	Nickel	0.1
Chloride	2.8	Nitrogen	10
Iron	2.8	Silicon	32
Sulphur	52.9	Sodium	1.0
Aluminium	0.6	Strontium	0.39
Barium	0.5	Tin	0.052
Berylium	0.02	Titanium	2.6
Bismuth	0.08	Tungsten	0.118
Calcium	35.6	Vanadium	12.0
Chromium	0.28	Zinc	0.126
Cobalt	0.01	Dysprosium	0.008
Indium	0.047	Europium	0.004
Lead	0.12	Gadolinium	0.005
Lithium	0.05	Samarium	0.004
Magnesium	0.088	Silver	0.02
Manganese	0.02		

Isotope	Calculated isotope activity in Bq/g in year 2005 WAGR Graphite				
	Moderator 110 t	**Top reflector 23t**	**Bottom reflector 23t**	**Side reflector 54t**	**Neutron Shield 73t**
Co-60	1.43E+03	4.57E+02	8.57E+02	9.76E+02	9.94E-01
Fe-55	7.35E+01	2.58E+01	5.09E+01	5.90E+01	3.88E-02
Ni-59	5.11E+03	1.83E+01	3.60E+00	4.17E+01	2.71E-02
Ni-63	4.72E+03	1.73E+03	3.39E+03	3.89E+03	2.59E+00
Mn-54	8.08E-07	2.21E-08	4.07E-04	5.31E-08	8.58E-11
Nb-94	4.34E-05	2.59E-06	3.54E-06	4.38E-06	1.29E-08
C-14	2.64E+04	9.37E+03	1.85E+04	2.14E+04	1.38E+01
H-3	7.79E+04	7.19E+04	7.80E+04	7.69E+04	2.60E+02
Eu-152	1.73E+00	9.22E+01	3.19E+00	2.01E+00	2.85E+01
Eu-154	2.59E+03	1.61E+03	2.36E+03	2.39E+03	1.62E+00
Eu-155	3.83E+02	1.65E+02	2.99E+02	3.11E+02	3.50E-02
Cl-36	4.87E+02	1.69E+02	3.47E+02	4.12E+02	2.55E-01
Ca-41	2.26E+02	7.24E+01	1.54E+02	1.84E+02	1.09E-01

Lesson Learnt

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Lesson Learnt

- **The main lesson learnt is do the minimum amount of work required to satisfy the regulatory bodies**